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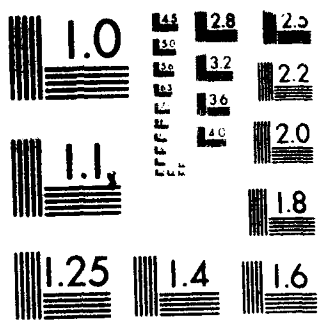

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I. INTRODUCTION

The research described in this final report involves the development of a new class of holographic materials. These materials exhibit two-photon photochemistry and thus are quadratic in their photochemical response to the intensity of the two recording laser beams. Unlike other previously suggested two-photon systems, the systems we have been developing can be used with milliwatts as opposed to megawatts of laser power. The holographic material consists of a photoactive organic molecule embedded in a polymer host. Grain free holograms can thus be recorded and no development or fixing step is necessary.

The quadratic nature of the photochemical response of the system offers several substantial advantages. First, consider the simple case where an attenuated beam is used to read the hologram. For a one-photon linear holographic material, the reading beam erases the hologram at a rate that is reduced from the writing rate by a factor that is equal to the ratio of the intensities of read and write beams. Two-photon systems respond, on the otherhand, to the square of this ratio. The deleterious effect of hologram erasure during reading is thus greatly reduced in these systems.

A second significant advantage is that in some cases it is possible to produce holograms in two-photon materials using two different wavelengths of light. The first wavelength can be viewed as switching on or gating the hologram formation process. It does not itself produce a hologram. The second wavelength, which does produce the hologram, may be in the infrared, making it possible to produce holograms that are compatible with GaAs, HeNe or Nd:YAG lasers. Furthermore, since no photochemistry is produced in the absence of the first wavelength, there is no erasure of the hologram during the read step. Since no light need be absorbed by the material during readout, this type of system is useful for applications in which high laser powers are used to read the hologram.

In this report, we summarize the work in this area that has been done under Contract DAAG29-82-C-0001. As described in the original proposal, work has focused on four different areas:

(1) Investigation of new two-photon holographic materials, particularly ones with higher maximum holographic efficiency.

(2) Demonstration of gating of the holographic recording process using an auxiliary light source.

(3) Development of two-photon holographic materials which respond to infrared laser wavelengths and their application in gated holography, and

(4) Production of simple holographic optical elements using these materials.

The next two sections provide a brief summary of the results obtained during the tenure of this contract. Section IV is a list of papers that have been or will be published which provide more of the technical details.

II. TECHNICAL REPORTS

Most holographic recording materials utilize photochemical reactions in which the rate of product formation depends linearly on laser light intensity. It can be shown that the overall rate constant k for product formation is proportional to the molar extinction coefficient ϵ and is linear in laser intensity I:

$$k \propto \epsilon I . \qquad (1)$$

Now consider using such a two-level medium for holographic recording. Such an experiment is shown schematically in Fig. 1. Normally the reconstruction wave will be at the same wavelength as the object and reference waves. This means that the reconstruction wave itself can also produce photochemistry. This photochemistry will result in gradual erasure of

the hologram. There are two ways of eliminating or minimizing the effect of erasure during reading for one-photon materials. The first, and less satisfactory method, is simply to reduce the reading light intensity so that the photochemistry that occurs during reading is reduced to an acceptable level. This is, of course, impractical when the hologram is meant to be semi-permanent. The most commonly used method of eliminating erasure is to fix the holographic image permanently. This is done in dichromated gelatin and silver halide films by wet chemical processes.

Another disadvantage of one-photon materials arises when one considers extending their sensitivity into the red and infrared regions. All of the energy used to produce photochemistry must come from the absorbed photons. Very few systems undergo efficient photochemistry in the red or infrared, and those few systems that do also tend to be thermally unstable.

Many of the problems with the one-photon systems are either not present or are significantly reduced in importance when the hologram forming photochemical process involves the absorption of two or more photons. Previous investigators have employed a two-photon three-level system. In a three-level system, the recording medium is simultaneously exposed to radiation at two different optical frequencies ω_1 and ω_2 with intensities I_1 and I_2. The rate constant k for product formation is now given by

$$k \propto \varepsilon_1 I_1 \varepsilon_2 I_2 \tau \qquad (2)$$

where ε_1 and ε_2 are the respective molar extinction coefficients at the two frequencies ω_1 and ω_2. τ is the lifetime of the intermediate state.

Consider the simple case where $\omega_1 = \omega_2$. The photochemistry is then proportional to I_1^2. Any reduction in the laser intensity during reading would have its effect on hologram

erasure reduced by the square of this factor. In addition, the energy to produce photochemistry is now supplied by two laser photons and it should not be difficult to find thermally stable two-photon systems that could be used with red or infrared radiation.

Perhaps a more interesting case is when $\omega_1 > \omega_2$. The medium is illuminated by reference and object beams at a frequency ω_2, as well as by an additional uniform and not necessarily coherent beam at ω_1. Holograms formed using this approach have several advantageous properties. As for the case where $\omega_1 = \omega_2$, these materials can be written with red or infrared lasers. Most of the energy needed to produce photochemistry is supplied by the ω_1 photon which can be in the ultraviolet. Another important advantage is that the recording medium is sensitive to the ω_2 radiation only when ω_1 radiation is present. Thus, during the reconstruction of the image for readout, no additional deleterious exposure of the recording medium takes place. Also, temporal or spatial gating of the recording can be achieved by masking or modulating the ω_1 beam.

The first demonstrations of two-photon holographic recording were in $LiNbO_3$ and KTN ($KTa_xNb_{1-x}O_3$), where the recording process was the multi-photon photorefractive effect. This effect involves the excitation of carriers within the crystal lattice to produce a macroscopic polarization change that induces a modulation of the index of refraction via the electro-optic effect. The major disadvantage of these materials is the lack of real intermediate energy levels between the ground and final states. Since the resonant enhancement from these intermediate levels is not present, the two-photon absorption processes are extremely weak and peak exposure intensities greater than 5 MW/cm^2 are necessary. A factor of 100 improvement in sensitivity was demonstrated by doping the $LiNbO_3$ crystal with Cr^{+3}, thereby converting the intermediate level from a virtual to a real

level with a 500 ns lifetime. The resulting sensitivity, however, was still several orders of magnitude too low for cw two-photon holography.

It can be shown that no three-level system can be sensitive enough to be used for holographic recording with cw lasers. This can be seen qualitatively by considering the role played by the intermediate state. For optimum production of photochemical products, one wants the population in this state to be as large as possible. This means that the extinction coefficient ε_1 should be large and the lifetime τ of this state should be long. However, these two quantities are not independent since lifetime and absorption strength are related to each other. There is thus an upper limit to the rate k that one can obtain for a three-level two-photon system.

To produce two-photon holographic systems that can be used with cw laser sources, one must circumvent these constraints. One way of doing this is to use a two-photon four-level system. In this case, there are now two intermediate levels. The level directly excited by radiation may have a large oscillator strength with respect to transitions from the ground state, while the second intermediate level may have a long lifetime. It thus should be possible to find a two-photon four-level system with which one can produce holograms using only a few milliwatts of cw laser power.

A variety of organic materials have such energy level systems. The first two states are ground state and an excited singlet state of the molecule, respectively. The other two states may be ground and excited states of an isomeric form of the molecule or they can be excited triplet states.

The holographic system that we have most thoroughly investigated in this context is biacetyl dissolved in a polymer host matrix. The relevant absorption spectra of biacetyl are

shown in Fig. 2. The initial photon excites the biacetyl molecule from its ground state to an excited singlet state S_n. This can be done according to the figure, provided the exciting wavelength is shorter than 500 nm. The second photon produces the hologram by exciting the biacetyl from $T_1 \rightarrow T_n$. This occurs if the second wavelength is between 600 and 1100 nm. The hologram is produced as a result of a photochemical reaction that occurs from T_n. The matrix in which the biacetyl was dissolved for these holographic experiments was a polycyanoacrylate polymer. Irradiation at ω_1 was provided in the UV by a medium pressure Hg-lamp. The infrared source at ω_2 was a Kr^+ laser operating at 752 nm. At time t=0, only the IR source illuminates the sample. Since this light is not absorbed by a ground state biacetyl molecule, no hologram is produced. When the UV source is turned on hologram formation begins. When it is turned off, the hologram growth is gated off. Holograms formed in this way require no chemical development step. They are "developed" simply by turning off the UV source.

We will next discuss several applications of four-level two-photon infrared holographic materials. The most familiar application for materials of this type is in the production of three-dimensional images. The recording of an extended image using the biacetyl/polycyanoacrylate system is shown in Fig. 3. The hologram was produced and reconstructed at 752.5 nm.

The advantage of recording extended holographic images in this way lies in the possibility of spatially or temporarily gating the process. By spatially gating hologram formation, one can imagine the production of holographic movies with frame to frame exposures gated on and off with the UV source. Temporal gating, of course, provides the potential for recording holograms at precise well-defined instants in time.

In the future, holograms will probably be most widely used for the production of a variety of kinds of optical elements for the deflection and focusing of light. Holographic optical elements (HOEs) utilize the fact that a hologram is capable of transforming a family of incident waves. The simples example of HOE is the holographic grating formed by the interference of plane object and reference waves. Another example is the holographic lens that is formed by the interference of a spherical object wave with a spherical or plane reference wave. Figure 4 shows the recording geometry for the production of an off-axis holographic lens.

The fundamental advantage of HOEs is that a complex and expensive optical system can be replaced by a cheap and simply fabricated piece of plastic. This is a particularly important consideration when one wishes to use the HOE with an inexpensive injection diode laser.

Figure 5 illustrates the results of the production of a holographic lens in the biacetyl/polycyanoacrylate system. The hologram was produced at 752.5 nm. Position 1 is near the lens focal point and Position 2 is beyond that point. One clearly sees the focusing effect of the HOE. The hologram was reconstructed at both 752.5 and 799.3 nm. It is clear in both cases that the HOE operates as a focusing element. It is also apparent, however, that the focal point is not as sharp for reconstruction at 799.3. This illustrates the importance point that the image quality and diffraction efficiency degrade as the reconstruction wavelength departs from the wavelength of the original object and reference waves.

As a final example of the use of the infrared hologram recording technique described above, we consider the fabrication of integrated optical devices. The general principal of these devices is illustrated in Fig. 6a. The two interfering infrared beams intersect inside a

narrow dielectric waveguide formed from a thin biacetyl/polycyanoacrylate layer between glass plates. Since the infrared beams are confined within a several micron thick dielectric layer, power densities can be quite high. Input powers of a few milliwatts can result in waveguide power densities of a few kilowatts/cm^2. This is particularly important for the two-photon four-level materials that tend to have low recording sensitivities.

A hologram can be formed at the intersection between the two infrared beams and a UV beam as shown in Fig. 6a. An example of an integrated optical device fabricated in this way is the directional coupler shown in Figs. 6b and 6c. In these photographs, one is looking down on the glass plates from above. The infrared light that is observed is scattered from defects in the recording layer. In Fig. 6c, the directional coupler is illustrated. A portion of the input beam is coupled by the device into another direction.

III. SUMMARY OF RESULTS

The work under this contract was directed in the four areas outlined in Section 1. The results in each of these areas are summarized below:

(1) Two photon holograms have been recorded at a variety of wavelengths with efficiencies that approach the theoretical limit of 70%.

(2) Gating has been observed in a variety of photochemical systems.

(3) The biacetyl photochemical system can be used for recording gated holograms with wavelengths in the near infrared.

(4) A variety of holographic optical elements have been produced using the biacetyl/polycyanoacrylate system.

The major area for future work is the improvement of the sensitivity of these two-photon four-level holographic systems. It still requires too much laser power for these

materials to be widely useful. We have examined many combinations of photochemically active molecule/polymer binder and have yet to find a better system than biacetyl/polycyanoacrylate.

IV. PUBLICATIONS

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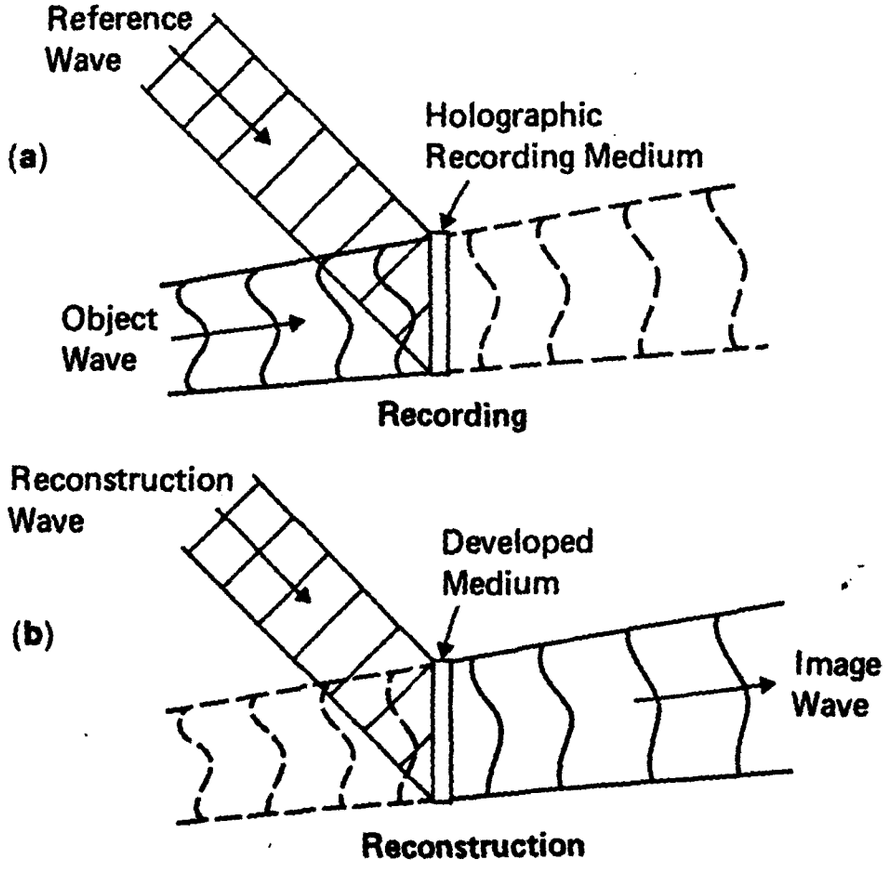

Reference
Wave
(a)
Holographic
Recording Medium
Object
Wave
Recording
Reconstruction
Wave
Developed
Medium
(b)
Image
Wave
Reconstruction

FIGURE 1

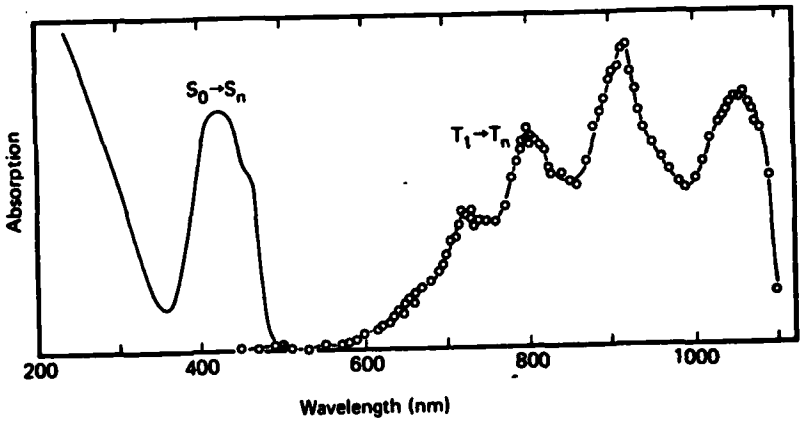
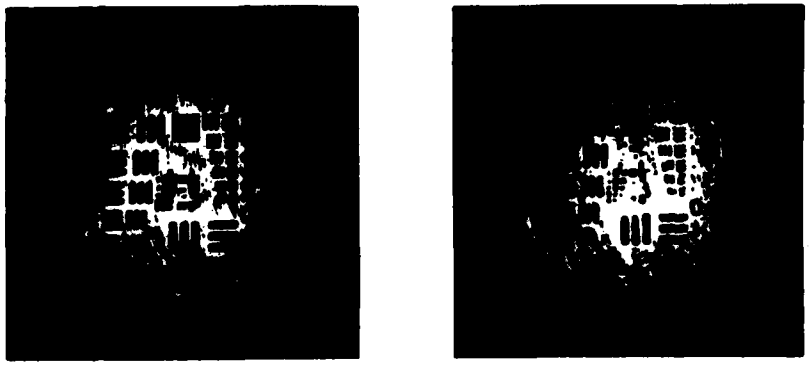

$S_0 \rightarrow S_n$
$T_1 \rightarrow T_n$
Absorption
200
400
600
800
1000
Wavelength (nm)

FIGURE 2

Two Photon Hologram



Original Reconstruction

FIGURE 3

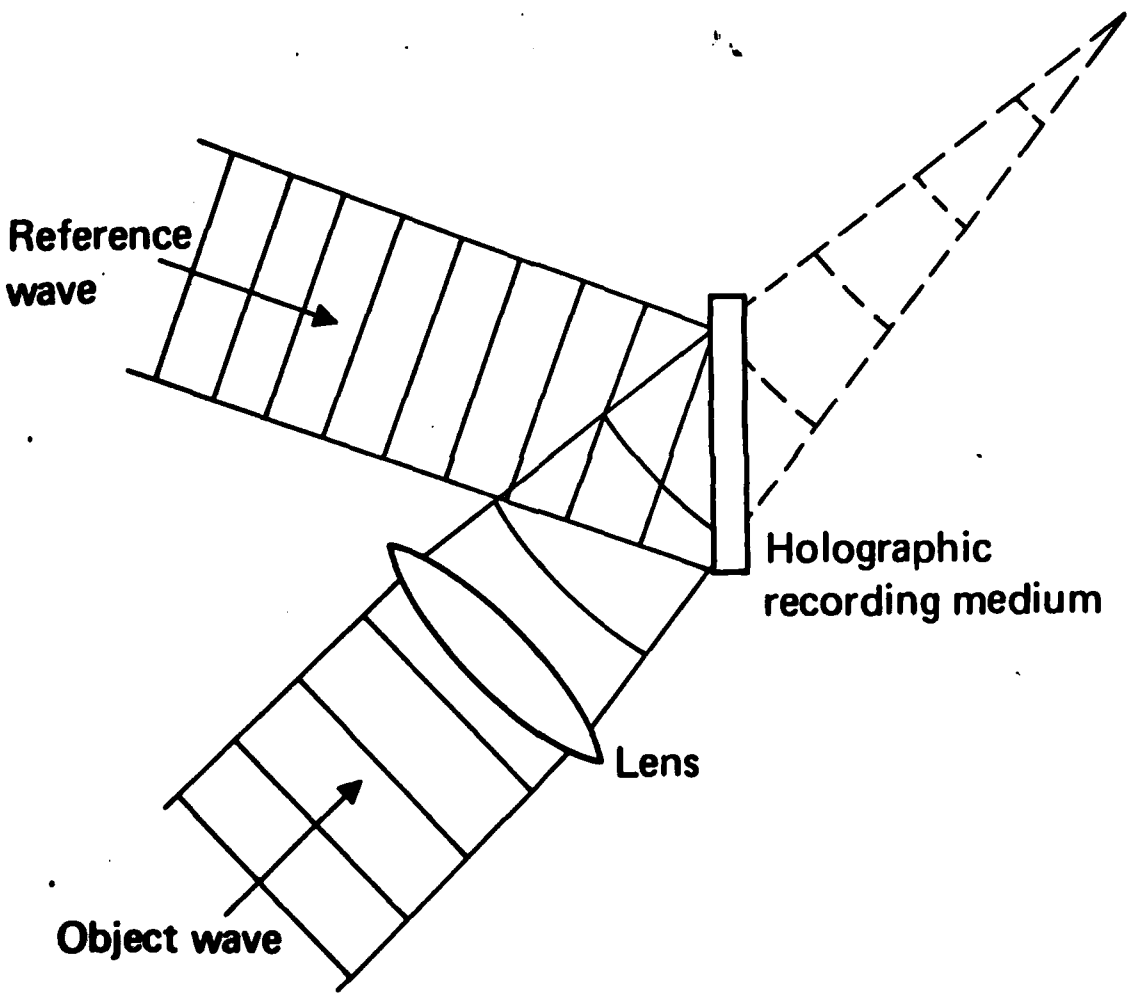

Reference
wave
Holographic
recording medium
Lens
Object wave

FIGURE 4

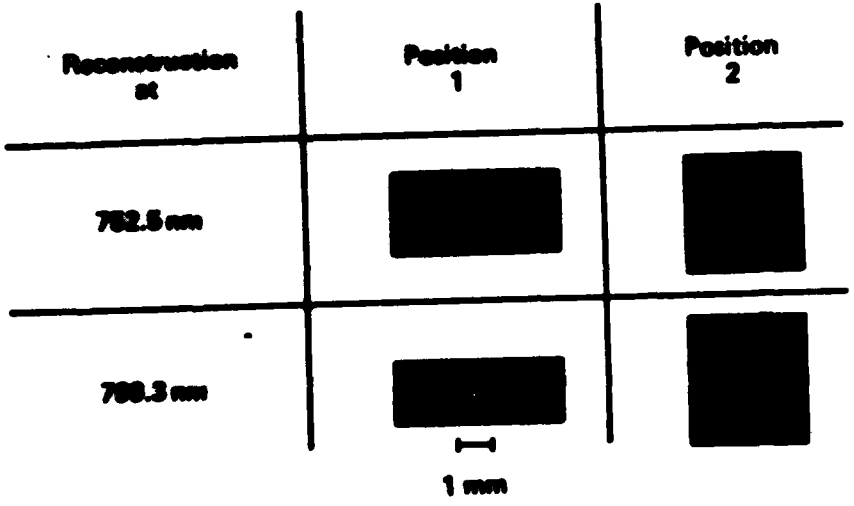


FIGURE 5

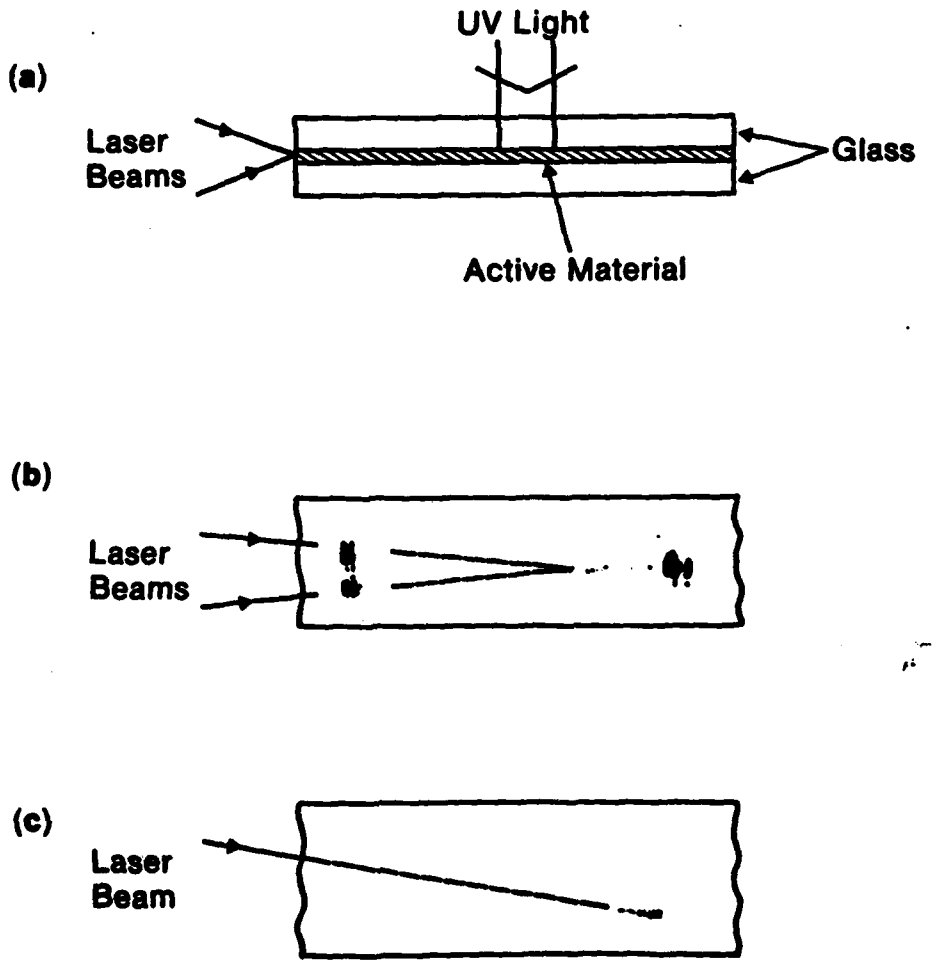

(a)
UV Light
Laser
Beams
Glass
Active Material
(b)
Laser
Beams
(c)
Laser
Beam

FIGURE 6